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GROWTH OF β BaB₂O₄ SINGLE CRYSTALS AND SHG EFFICIENCY MEASUREMENTS

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Abstract - We report here the growth and characterization of non linear single crystals β BaB₂O₄ (BBO) which is an interesting material for second harmonic generation. Single crystals were grown from Na₂O-B₂O₃ solutions using the top seeded solution growth method. The experimental set up included a Nd:YAG, Q switched, focalised with a cylindrical lens on the samples. Efficiency up to 40 % were obtained using 6 mm long crystal.

1. - Introduction

β BaB₂O₄ (BBO) is an interesting material for second harmonic generation (SHG) (1). It exhibits high damage threshold and a wide transparency range (2). BaB₂O₄ is known to exist in two phases : a high temperature α phase and a low temperature β phase with a transition temperature of about 925° C (3). α BaB₂O₄ melts congruently at 1096° C but the growth of β phase requires a growth temperature below 925° C in a solvent. The real success in growing β BaB₂O₄ came with discovery of a suitable solvent. The workers of Fujian Institute have studied all of the practical solvent for this material : BaCl₂, BaF₂, Li₂O, NaBO₂ and Na₂O (4). They reported that Na₂O yields the best results in terms of crystal size and defect density.

2. - Crystal growth procedure**2.1 - Preparation of charges**

Several melt compositions were investigated in the system BBO-Na₂O-B₂O₃. In order to obtain the desired low temperature β phase, growth must be carried out from solution below 925° C. With Na₂O-B₂O₃ as solvent, we find that β BaB₂O₄ can be crystallized at about 900° C from solutions with typical composition of 80 % mole BBO - 16 % mole Na₂O - 4 % mole B₂O₃. Charges were weighed out and fractionnal amounts were placed in pure platinum crucible to be melted in a synthesis furnace at 1025° C.

2.2 - Top seeded solution growth apparatus

High thermal gradient furnace was used in order to initiate a good growth from the seeds and to minimize the defects density in the crystals. At the melt surface the axial temperature gradient was about 50° C/cm. BBO seeds were held rigidly using a platinum holder. Before seeding the melt was heated at about 1000° C during several hours.

2.3 - Growth conditions

Crystal growth experiments were performed with the use of seeds oriented along the y-axis. BBO seeds having a rotation rate of 10-25 rpm were introduced carefully in the melt in order to avoid nucleation, at about 900° C to minimize solution viscosities. Typically we used a cooling rate of 0.025° C/hour and a pulling rate of 1 mm/day.

3. - Results

BBO crystals boules of 40 mm in diameter and 10 mm in thickness were grown. The crystals had tendency to fracture along the basal plane during cooling. Most crystals grown presented internal defects, typically : solvent inclusions, fractures and cleavages.

4. - Characterization

After growth, crystals were characterized by optical microscopy in order to localise the defects. Following these observations, we prepared single parallelepipedic crystals (5 x 5 x 8 mm³) of good optical quality, to make the SHG efficiency measurements. Two plane and parallele faces were then oriented and polished. Two orientations were studied : one perpendicular to the optical axis and one at 22°8 of this axis corresponding with the type I phase matching for SHG at $\lambda = 1064$ nm.

The spectral transparency range was measured using a spectrophotometer and found to spread from 200 nm to 2500 nm (at a level of 1/2) (figure 1).

FIG. 1 - Transmission of β BaB₂O₄ crystal

5. - Second harmonic generation experiments

5.1 - Crystal orientation

The low temperature phase for baryum metaborate can be indexed with a monoclinique cell where $a = 11.133 \text{ \AA}$, $b = 12.67 \text{ \AA}$, $c = 8.381 \text{ \AA}$ and $\beta = 100.03^\circ$ (5). In this representation, the phase matching direction for the second harmonic generation ($22^\circ 8'$ from the optical axis) is not perpendicular to a common crystallographic plane (6). So the problem consisted to determine this SHG plane. Thus, we found two crystallographic planes, (010) and (201), which are both perpendicular to the SHG plane. The orientation was realized with the Laue method. Only one pattern was required, with the software "*Orient Express*" to locate accurately the crystallographic planes (7). When these two (010) and (201) orientations are located, we cut and polish, perpendicular to them, two parallele faces.

5.2 - Results

The SHG measurements were made using as the fondamental beam source a Q-Switched Nd:YAG having the following properties :

- $\lambda = 1,064 \text{ }\mu\text{m}$
- $\nu = 1 \text{ KHz}$
- $\tau = 17 \text{ ns}$
- $E_{\text{pulse}} = 50 \text{ mJ}$ linearly polarized around 80 % (so the usefull energy was 40 mJ)
- beam quality = 3 times the diffraction limit

The beam was focused using a cylindrical lens and its size in the BBO crystal was (FWHM) : $290 \times 4 \text{ }\mu\text{m}^2$ leading to a power density (FWHM) of $0,27 \text{ GW/cm}^2$. With this experimental set up and using a 6 mm long BBO crystal in the type I phase matching configuration, an energy of 17 mJ was generated at $\lambda = 532 \text{ nm}$ which gives a conversion efficiency of 40 %.

6. - Conclusion

Single crystals of $\beta \text{ BaB}_2\text{O}_4$ have been grown from $\text{Na}_2\text{O-B}_2\text{O}_3$ solutions. High thermal gradient furnace was used. Crystals grown from "y" oriented seed had tendency to fracture along the basal plane and to contain some optical defects. Parallelepipedic samples ($5 \times 5 \times 8 \text{ mm}$) have been prepared and tested for second harmonic generation efficiency. Values up to 40 % were obtained. Crystal growth with "z" oriented seeds are presently in progress.

7. - Acknowledgements

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